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## Citation

Hamidian, M. H., S. D. Edkins, Chung Koo Kim, J. C. Davis, A. P. Mackenzie, H. Eisaki, S. Uchida, et al. 2015. "Atomic-Scale Electronic Structure of the Cuprate d-Symmetry Form Factor Density Wave State." *Nature Physics* 12 (2) [October 26]: 150–156. doi:10.1038/nphys3519.

## Published Version

doi:10.1038/nphys3519

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# Atomic-scale Electronic Structure of the Cuprate *d*-Symmetry Form Factor Density Wave State

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## ABSTRACT

Extensive research into high temperature superconducting cuprates is now focused upon identifying the relationship between the classic ‘pseudogap’ phenomenon<sup>1,2</sup> and the more recently investigated density wave state<sup>3-13</sup>. This state always exhibits wavevector  $Q$  parallel to the planar Cu-O-Cu bonds<sup>4-13</sup> along with a predominantly *d*-symmetry form factor<sup>14 - 17</sup> (dFF-DW). Finding its microscopic mechanism has now become a key objective<sup>18-30</sup> of this field. To accomplish this, one must identify the momentum-space ( $k$ -space) states contributing to the dFF-DW spectral weight, determine their particle-hole phase relationship about the Fermi energy, establish whether they exhibit a characteristic energy gap, and understand the evolution of all these phenomena throughout the phase diagram. Here we use energy-resolved sublattice visualization<sup>14</sup> of electronic structure and show that the characteristic energy of the dFF-DW modulations is actually the ‘pseudogap’ energy  $\Delta_1$ . Moreover, we demonstrate that the dFF-DW modulations at  $E=-\Delta_1$  (filled states) occur with relative

phase  $\pi$  compared to those at  $E=\Delta_1$  (empty states). Finally, we show that the dFF-DW  $Q$  corresponds directly to scattering between the ‘hot frontier’ regions of  $k$ -space beyond which Bogoliubov quasiparticles cease to exist<sup>31,32,33</sup>. These data demonstrate that the dFF-DW state is consistent with particle-hole interactions focused at the pseudogap energy scale and between the four pairs of ‘hot frontier’ regions in  $k$ -space where the pseudogap opens.

A conventional ‘Peierls’ charge density wave (CDW) in a metal results from particle-hole interactions which open an energy gap at specific regions of  $\mathbf{k}$ -space that are connected by a common wavevector  $\mathbf{Q}$ . This generates a modulation in the density of free charge at  $\mathbf{Q}$  along with an associated modulation of the crystal lattice parameters. Such CDW states are now very well known<sup>34</sup>. In principle, a density wave modulating at  $\mathbf{Q}$  can also exhibit a ‘form factor’ (FF) with different possible symmetries<sup>35,36</sup> (see Supplementary Information (SI) Section 1). This is relevant to the high-temperature superconducting cuprates because numerous researchers have recently proposed that the ‘pseudogap’ regime<sup>1,2</sup> (PG in Fig. 1A) contains an unconventional density wave with a  $d$ -symmetry form factor<sup>18-30</sup>. The basic phenomenology of such a state is that intra-unit-cell (IUC) symmetry breaking renders the  $O_x$  and  $O_y$  sites within each  $\text{CuO}_2$  unit-cell electronically inequivalent, and that this inequivalence is then modulated periodically at wavevector  $\mathbf{Q}$  parallel to  $(1,0);(0,1)$ . The real-space ( $\mathbf{r}$ -space) schematic of such a  $d$ -symmetry FF density wave (dFF-DW) at  $\mathbf{Q}_x$  as shown in Fig. 1B exemplifies periodic modulations at the  $O_x$  sites that are  $\pi$  out of phase with those at the  $O_y$  sites. This state is then described by  $A(\mathbf{r}) = D(\mathbf{r})\cos(\phi(\mathbf{r}) + \phi_0(\mathbf{r}))$ , where  $A(\mathbf{r})$  represents the modulating electronic degree of freedom,  $\phi(\mathbf{r}) = \mathbf{Q}_x \cdot \mathbf{r}$  is the DW spatial phase at location  $\mathbf{r}$ ,  $\phi_0(\mathbf{r})$  represents disorder related spatial phase shifts, and  $D(\mathbf{r})$  is the magnitude of the  $d$ -symmetry form factor<sup>14,22,24</sup>. To distinguish between the various microscopic mechanisms proposed for the  $\mathbf{Q}=(Q,0);(0,Q)$  dFF-DW state of cuprates<sup>18-30</sup>,

it is essential to establish its atomic-scale phenomenology, including the  $\mathbf{k}$ -space eigenstates contributing to its spectral weight, the relationship (if any) between modulations occurring above and below the Fermi energy, whether the modulating states in the DW are associated with a characteristic energy gap, and how the dFF-DW evolves with doping.

To visualize such phenomena directly as in Fig. 1C, we use sublattice phase-resolved imaging of the electronic structure<sup>14</sup> of the  $\text{CuO}_2$  plane. Both the scanning tunneling microscope (STM) tip-sample differential tunneling conductance  $\frac{dI}{dV}(\mathbf{r}, E = eV) \equiv g(\mathbf{r}, E)$  and the tunnel-current  $I(\mathbf{r}, E)$  are measured at bias voltage  $V=E/e$  and with sub-unit-cell spatial resolution. Because the density-of-electronic-states  $N(\mathbf{r}, E)$  is related to the differential conductance as  $g(\mathbf{r}, E) \propto \left[ eI_s / \int_0^{eV_s} N(\mathbf{r}, E') dE' \right] N(\mathbf{r}, E)$  with  $I_s$  and  $V_s$  being arbitrary parameters and the denominator  $\int_0^{eV_s} N(\mathbf{r}, E') dE'$  unknown, valid determination of  $N(\mathbf{r}, E)$  is impossible (SI Section 2). However, one can suppress these serious systematic errors by using  $R(\mathbf{r}, E) = I(\mathbf{r}, E)/I(\mathbf{r}, -E)$  or  $Z(\mathbf{r}, E) = g(\mathbf{r}, E)/g(\mathbf{r}, -E)$ ; this allows distances, wavelengths, and spatial-phases of electronic structure to be measured accurately. The unprocessed  $g(\mathbf{r}, E)$  acquired for and analyzed in this paper: (i) are measured over very large fields of view (to achieve high phase-precision in Fourier analysis) ; (ii) simultaneously maintain deeply sub-unit-cell precision measurements in  $\mathbf{r}$  (to achieve high precision in sublattice segregation); (iii) are taken over a wide range of energies  $E$  with fine energy-spacing, so that energy dependences of  $d$ -symmetry FF modulations may be accurately determined. We then calculate each sublattice-phase-resolved  $Z(\mathbf{r}, E)$  image and separate it into three: the first,  $\text{Cu}(\mathbf{r})$ , contains only the measured values of  $Z(\mathbf{r})$  at Cu sites while the other two,  $O_x(\mathbf{r})$  and  $O_y(\mathbf{r})$ , contain only the measurements at the  $x/y$ -axis oxygen sites. Phase-resolved Fourier transforms of the  $O_x(\mathbf{r})$  and  $O_y(\mathbf{r})$  sublattice images<sup>14</sup>,



$\tilde{O}_x(\mathbf{q}) = \text{Re}\tilde{O}_x(\mathbf{q}) + i\text{Im}\tilde{O}_x(\mathbf{q})$ ;  $\tilde{O}_y(\mathbf{q}) = \text{Re}\tilde{O}_y(\mathbf{q}) + i\text{Im}\tilde{O}_y(\mathbf{q})$ , are used to determine the form factor symmetry for modulations at any  $\mathbf{q}$

$$\tilde{D}^Z(\mathbf{q}) = (\tilde{O}_x(\mathbf{q}) - \tilde{O}_y(\mathbf{q}))/2 \quad (1a)$$

$$\tilde{S}'^Z(\mathbf{q}) = (\tilde{O}_x(\mathbf{q}) + \tilde{O}_y(\mathbf{q}))/2 \quad (1b)$$

$$\tilde{S}^Z(\mathbf{q}) = \tilde{C}u(\mathbf{q}) \quad (1c)$$

where the superscript Z identifies the type of sublattice resolved data used. Specifically for a DW occurring at  $\mathbf{Q}$ , one can then evaluate the magnitude of its  $d$ -symmetry form factor  $\tilde{D}(\mathbf{Q})$  and its  $s'$ - and  $s$ -symmetry form factors  $\tilde{S}'(\mathbf{Q})$  and  $\tilde{S}(\mathbf{Q})$ , respectively. Studies of non-energy-resolved  $R(\mathbf{r}, E)$  images using this approach have revealed that the DW modulations in the  $O_x(\mathbf{r})$  and  $O_y(\mathbf{r})$  sublattice images of electronic structure in underdoped  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$  and  $\text{Ca}_{2-x}\text{Na}_x\text{CuO}_2\text{Cl}_2$  consistently exhibit a relative phase of  $\pi$  and therefore a  $d$ -symmetry form factor<sup>14</sup>; X-ray scattering studies<sup>15,16</sup> yield the same conclusion for two other cuprates,  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  and  $\text{Bi}_2\text{Sr}_{2-x}\text{La}_x\text{CuO}_{6+x}$ .

X-ray scattering studies report a short-ranged density wave with wavevector centered around  $\mathbf{Q}=(Q,0);(0,Q)$  occurring approximately in the pink shaded regions<sup>11,12,13</sup> of the schematic phase diagram in Fig 1A. Figures 1B,C exemplify the predominately  $d$ -symmetry form factor<sup>14-17</sup> of this DW when imaged directly. One obstacle to understanding this dFF-DW state is that large field-of-view sublattice-resolved images of cuprate electronic structure<sup>14</sup> never look like an ideal long-range ordered version of Fig. 1B. Instead, Fig. 2A shows a typical  $Z(\mathbf{r}, 150\text{meV})$  image of  $p=8\%$   $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$ , for  $T \ll T_c$  in the superconducting phase while Fig. 2B shows the equivalent  $Z(\mathbf{r}, 150\text{meV})$  for  $T > T_c$  in the cuprate pseudogap phase. While elements indistinguishable from Fig. 1C can be seen in 2A, B, no long range order is obvious. Therefore, to explore the spatial arrangements of the dFF-DW in such electronic structure images, we analyze  $\tilde{D}^Z(\mathbf{q})$  which is a robust sublattice-phase-resolved measure of the  $d$ -symmetry form factor (SI Section 3). Analysis of Figures 2A, B in this

fashion yields  $2C,D$ ; both clearly exhibit the dFF-DW maxima at the two inequivalent wavevectors  $\mathbf{Q}_x$  and  $\mathbf{Q}_y$ . Fourier filtering these two  $\tilde{D}^Z(\mathbf{q})$  from  $2A,B$  for only those regions surrounding  $\mathbf{Q}_x$  and  $\mathbf{Q}_y$  (within dashed circles) generates two complex-valued images  $D_x(\mathbf{r}), D_y(\mathbf{r})$

$$D_x(\mathbf{r}) = \frac{2}{(2\pi)^2} \int d\mathbf{q} e^{i\mathbf{q}\cdot\mathbf{r}} \tilde{D}^Z(\mathbf{q}) e^{-\frac{(\mathbf{q}-\mathbf{Q}_x)^2}{2\Lambda^2}} ; D_y(\mathbf{r}) = \frac{2}{(2\pi)^2} \int d\mathbf{q} e^{i\mathbf{q}\cdot\mathbf{r}} \tilde{D}^Z(\mathbf{q}) e^{-\frac{(\mathbf{q}-\mathbf{Q}_y)^2}{2\Lambda^2}} \quad (2a)$$

where  $\Lambda^{-1}$  is the characteristic length scale over which variations in  $D_x(\mathbf{r}), D_y(\mathbf{r})$  can be resolved, set by the filter width in Fourier space.

Their magnitudes

$$|D_x(\mathbf{r})| = \sqrt{(\text{Re}D_x(\mathbf{r}))^2 + (\text{Im}D_x(\mathbf{r}))^2} \quad (2b)$$

$$|D_y(\mathbf{r})| = \sqrt{(\text{Re}D_y(\mathbf{r}))^2 + (\text{Im}D_y(\mathbf{r}))^2} \quad (2c)$$

represent the local amplitudes of dFF-DW modulations along  $\mathbf{Q}_x$  and  $\mathbf{Q}_y$ , respectively. Any unidirectional domain arrangements of the dFF-DW state can then be determined by introducing

$$F(\mathbf{r}) = \frac{|D_x(\mathbf{r})| - |D_y(\mathbf{r})|}{|D_x(\mathbf{r})| + |D_y(\mathbf{r})|} \quad (3)$$

which is designed to identify regions where the dFF-DW modulation is primarily along the x-axis or the y-axis depending on the sign of  $F(\mathbf{r})$  (SI Section 4). Figures 2E,F show how regions of  $-1.0 < F(\mathbf{r}) < -0.3$  (shaded blue) are primarily modulating along y-axis while regions  $+0.3 < F(\mathbf{r}) < +1.0$  (shaded orange) are primarily modulating along x-axis (those with  $-0.3 < F(\mathbf{r}) < +0.3$  shaded white appear at boundaries). Figures 2G and 2H reveal the results of this analysis for the data in Figs 2A,B respectively. Overall, the system is configured into spatial regions within which the dFF-DW along one direction is dominant. By overlaying the color scale for  $F(\mathbf{r})$  on the data in Figs 2A,B to create Figs 2G,H, one can see directly the unidirectional region configurations derived from Eqn. 3.

These observations of coexisting nanoscale unidirectional regions are in reasonable agreement with related X-ray studies<sup>37</sup> of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ . Finally, since the data in Fig. 2B and Fig. 2H were measured at  $T > T_c$  (pink region Fig. 1A), it demonstrates directly that the cuprate dFF-DW appears first in the non-superconducting ‘pseudogap’ regime.

A conventional CDW state opens a gap in the energy spectrum of  $\mathbf{k}$ -space electronic eigenstates with the maximum spectral weight of modulating states occurring at the edges of this energy-gap<sup>34</sup>. But which energy gap (if any) is associated with the dFF-DW state found in underdoped cuprates is unknown. Figure 3A shows how a typical tunneling conductance spectrum representative of strongly underdoped cuprates exhibits two characteristic energies<sup>31,32,33</sup>. While the lower energy scale  $\Delta_0$  represent the maximum energy at which Bogoliubov quasiparticle excitations exist<sup>31,32,33</sup> (see Figure 3B) the higher energy scale (dashed blue line) is the cuprate ‘pseudogap’ as determined from its comparison with doping dependence of pseudogap scale in tunneling and photoemission. To identify the energy dependence of the cuprate dFF-DW states, we measure  $Z(\mathbf{r}, |E|)$  and from it calculate  $\tilde{D}^Z(\mathbf{q}, |E|)$ ,  $\tilde{S}'^Z(\mathbf{q}, |E|)$ , and  $\tilde{S}^Z(\mathbf{q}, |E|)$ . Figure 3C shows the measured power-spectral-density of the  $d$ -symmetry FF modulations  $\tilde{D}^Z(\mathbf{q}, |88\text{meV}|)^2$ , with the wavevectors near  $\mathbf{Q}_x$  and  $\mathbf{Q}_y$  indicated by red circles. Adopting the common convention in X-ray studies<sup>9,10,11,16</sup> for determining the DW wavevector magnitude  $|\mathbf{Q}|$ , we measure  $\tilde{D}^Z(\mathbf{q}, |E|)$  along a line in the high symmetry directions (1,0):(0,1) passing through the region of the dFF-DW peak and fit these data to a background plus Gaussian. The peak positions of the two Gaussians are then assigned to be the values of  $\mathbf{Q}_x$  and  $\mathbf{Q}_y$  (although a broad range of values of  $\mathbf{Q}$  can actually be detected under each peak in  $\tilde{D}^Z(\mathbf{q}, |E|)$  e.g. dashed circles Figs 2C,D). It remains to be determined whether these incommensurate maxima in  $\tilde{D}^Z(\mathbf{q}, |E|)$  are due to domains of continuously incommensurate dFF-DW or domains of commensurate dFF-DW separated by discommensurations<sup>38</sup>. Nevertheless, Figure 3B plots the energy

dependence of the dFF-DW wavevectors (blue line) determined in this way for a  $p = 0.06$  sample. Such information was not previously available from measurements of the modulation wavevectors from STM images lacking sublattice phase-resolved segregation<sup>14</sup> into  $\tilde{O}_x(\mathbf{q})$  and  $\tilde{O}_y(\mathbf{q})$ . Figure 3E shows the measured  $\mathbf{k}$ -space locus where Bogoliubov quasiparticles exist<sup>31,32,33</sup> as a function of hole density. When the dispersive ‘octet’ of Bogoliubov scattering interference disappears a transition occurs to a non-dispersive density wave modulation (Fig. 3B). In Fig. 3D, the doping dependence of the conventional  $\mathbf{Q}_x$ ,  $\mathbf{Q}_y$  of the  $d$ -symmetry form factor modulations is shown using blue symbols, while the measured shortest wavevectors interconnecting the  $\mathbf{k}$ -space arc tips (Fig. 3E) are indicated by using colored symbols referring to each hole-density in Fig. 3E. These data demonstrate directly that the conventional  $\mathbf{Q}_x$ ,  $\mathbf{Q}_y$  of the dFF-DW state corresponds to the locations in  $\mathbf{k}$ -space of arc tips. Finally, in Fig. 3F we show the measured energy dependences of the amplitudes of the  $s$ ,  $s'$ - and  $d$ -form factor modulations,  $S^Z(E)$ ,  $S'^Z(E)$  and  $D^Z(E)$ , determined by integrating over the region of  $\mathbf{q}$ -space enclosed by solid red circles in 3C (SI Section 5). The  $d$ -symmetry form factor is negligible for modulations in the low energy range that contains only Bogoliubov quasiparticles (and which we now see is dominated by  $s'$ -symmetry form factors) but it rapidly becomes intense at higher energy and reaches maximum at the pseudogap energy scale which for this sample is  $\Delta_1 \sim 90\text{meV}$ . This reveals that the characteristic energy of electronic-structure modulations in the cuprate  $d$ -symmetry FF density wave is actually the pseudogap energy.

As a function of energy, the transition from Bogoliubov quasiparticle interference modulations to dFF density wave modulations occurs in an unusual fashion. Although Bogoliubov QPI is observed as expected everywhere on the Fermi surface in overdoped cuprates<sup>33</sup>, in underdoped samples it evolves as expected only until the energy  $E \approx \Delta_0$  at which the terminations of the Bogoliubov coherent  $\mathbf{k}$ -space arcs (Fig. 3E)

are observed<sup>31,32,33</sup>. Here, the set of seven dispersive scattering interference modulations  $\mathbf{q}_1, \mathbf{q}_2, \dots, \mathbf{q}_7$  signifying Bogoliubons<sup>32</sup> (SI Section 5) disappears in a narrow energy window during which dispersion of the two surviving modulations  $q_1(E)$  and  $q_5(E)$  comes to a halt, leaving the non-dispersive dFF-DW modulations with  $\mathbf{q}_1^* \approx \mathbf{q}_1(\Delta_0)$  and  $\mathbf{q}_5^* \approx \mathbf{q}_5(\Delta_0)$  (see Fig. 3B and Refs. 31,32,33). The intensity of these modulations first becomes detectable at  $\Delta_0$  and, as we show below, reaches an intense maximum at  $\Delta_1$ , all the while maintaining the same wavevectors  $\mathbf{Q}_x^d$  and  $\mathbf{Q}_y^d$  as shown in Fig. 3B. We refer to this  $\mathbf{k}$ -space region where Bogoliubov quasiparticles yield to modulations of a dFF-DW as the ‘hot frontier’<sup>39</sup> to distinguish it from the colloquial ‘hot spots’ beyond which, in a conventional density wave, dispersive quasiparticle states would reappear. In cuprates, this does not occur and, instead, the ‘hot frontiers’ define the  $\mathbf{k}$ -space limit beyond which only dFF-DW modulations are detected<sup>31,32,33</sup> using SI-STM (blue in Fig. 3B).

Key information on the microscopic cause of any DW state is also contained in the relationship between modulations of states above and below the Fermi energy. For example, mixing via interactions of states with momenta  $\mathbf{k}_1$  and  $\mathbf{k}_2$  generates modulations at wavevector  $\mathbf{Q} = \mathbf{k}_1 - \mathbf{k}_2$ . The wavefunctions of any resulting DW would then form bonding/anti-bonding states below/above the Fermi level which are proportional to  $e^{i\mathbf{k}_1 \cdot \mathbf{r}} \pm e^{i\mathbf{k}_2 \cdot \mathbf{r}}$ . The related densities of these states would then exhibit modulations governed by  $|e^{i\mathbf{k}_1 \cdot \mathbf{r}} \pm e^{i\mathbf{k}_2 \cdot \mathbf{r}}|^2 = 2(1 \pm \cos(\mathbf{Q} \cdot \mathbf{r}))$ . In such scenarios, the DW modulations above the Fermi energy should always be  $\pi$  out-of-phase with the equivalent ones below. To explore this issue in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$ , we show in Figs 4A,B the measured  $g(\mathbf{r}, +94\text{meV})$  from filled states and  $g(\mathbf{r}, -94\text{meV})$  from empty states, respectively, each at the characteristic energy of the dFF-DW (Fig 3F and SI Section 6). For these two images the sublattice-phase-resolved  $\tilde{D}^g(\mathbf{q}, E)$  (Eqn. 1a) are calculated and reveal a predominantly  $d$ -symmetry form factor modulation with wavevectors near

$\mathbf{Q}_x$  and  $\mathbf{Q}_y$  in Figs 4A,B. Next, by Fourier filtering these two  $\tilde{D}^g(\mathbf{q}, E)$  for regions surrounding  $\mathbf{Q}_y$  we determine the complex-valued  $D_y(\mathbf{r})$  and thus the spatial phase of dFF-DW modulation along  $\mathbf{Q}_y$  as

$$\phi_y(\mathbf{r}, E) = \arctan \left( \text{Im} D_y(\mathbf{r}, E) / \text{Re} D_y(\mathbf{r}, E) \right) \quad (4a)$$

For  $E=+94\text{meV}$  this is shown in Fig. 4C and for  $E=-94\text{meV}$  in Fig. 4D. Visual comparison reveals that these two  $\phi_y(\mathbf{r}, \pm E)$  images are out of phase with each other by  $\pi$ . And indeed, the spatial-average value of  $\phi_y(\mathbf{r}, +E) - \phi_y(\mathbf{r}, -E)$  as a function of  $E$  (over the whole field of view A and B) is shown in Fig. 4F. It reveals that, while the relevant  $\mathbf{Q}_x$  and  $\mathbf{Q}_y$  components of  $g(\mathbf{r}, +E)$  and  $g(\mathbf{r}, -E)$  images are in phase with each other at low energy, they rapidly evolve at  $|E| > 70\text{meV}$  and become globally  $\pi$  out of phase at  $|E| \sim \Delta_1$  (Figs 4A,B). The shaded region indicates evolution through a situation where some areas exhibit  $\phi \sim 0$  and some  $\phi \sim \pi$  but this is quickly resolved upon reaching pseudogap energy  $\Delta_1$ . Similar analysis for the particle-hole symmetry in phases  $\phi_x(\mathbf{r}, \pm E)$  of  $\mathbf{Q}_x$  modulations

$$\phi_x(\mathbf{r}, E) = \arctan \left( \text{Im} D_x(\mathbf{r}, E) / \text{Re} D_x(\mathbf{r}, E) \right) \quad (4b)$$

yields a virtually identical result. These phenomena also occur throughout the underdoped regions the phase diagram (SI Section 6.III). All these data demonstrate that a phase difference of  $\pi$  exists between spatial modulations of the filled states at pseudogap energy  $E \sim -\Delta_1$  and the empty states at  $E \sim +\Delta_1$ , for the cuprate dFF-DW state.

To summarize: by introducing new techniques to determine the energy/momentum and doping dependence of modulation form factor symmetry, we find that the predominantly  $d$ -symmetry form factor density wave exists throughout the underdoped region of the  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  phase diagram (Fig. 3D) including in the pseudogap regime  $T > T_c$  (Figs 1C, 2B). The spatial arrangements are primarily in the form of nanoscale regions each containing a primarily unidirectional dFF-DW (Figs

2G,H). The conventionally defined wavevectors  $\mathbf{Q}_x$  and  $\mathbf{Q}_y$  of the dFF-DW state evolve with doping as determined by the four shortest scattering vectors linking the  $\mathbf{k}$ -space regions beyond which Bogoliubov quasiparticle excitations are nonexistent (Figs 3D,E) and at which the pseudogap emerges. Further, we demonstrate that, as determined in terms of tunneling probabilities, the dFF-DW state is particle-hole antisymmetric in the sense that a phase difference of  $\pi$  exists between spatial modulations of the filled states ( $E \sim -\Delta_1$ ) and the empty states ( $E \sim +\Delta_1$ ) (Fig. 4E). Most significantly, we show that the characteristic energy of the dFF-DW electronic-structure modulation is actually the pseudogap energy  $\Delta_1$  (Fig. 3F).

These data provide evidence that the cuprate  $d$ -symmetry form factor density wave state involves particle-hole interactions, and that these occur close to wavevectors interconnecting the ‘hot frontiers’ in  $\mathbf{k}$ -space at which the pseudogap emerges<sup>31,32,33</sup>. Moreover, the dFF-DW electronic structure modulations have a characteristic energy scale which is the pseudogap energy. This intimate connection of the dFF-DW state with the pseudogap electronic structure is consistent with the fact that this state is only found within the pseudogap regime<sup>11-13</sup>. Of course, electron-lattice interactions can also play a significant role, with the coupling to the  $B_{1g}$  modes long being of foremost interest<sup>20,21,40</sup>. Strong interactions of this mode with the electrons<sup>41</sup> ultimately leading to static, finite  $\mathbf{Q}$ , lattice distortions with  $d$ -symmetry form factor<sup>42</sup> have recently been discovered in association with the cuprate dFF-DW state. Nevertheless, electron-lattice interactions are not by themselves sufficient to explain the phase diagram of the dFF-DW<sup>11-13</sup> because, for example, they also exist in the overdoped regime where the dFF-DW is absent. Moreover, theoretical models involving  $\mathbf{k}$ -space instabilities<sup>27,28,30,43</sup> which are consistent with the results herein, emphasize that a density wave with this  $\mathbf{Q}$  and form factor symmetry cannot emerge from a large Fermi surface; instead, a preexisting reorganization of  $\mathbf{k}$ -space due to the pseudogap would be required. Overall, our data

support a microscopic picture in which the exotic electronic structure of the pseudogap is parent to the  $dFF$ -DW state and not vice-versa, where the energy-scale and wavevectors of the  $dFF$ -DW are intimately linked to those of the pseudogap, and in which the  $dFF$ -DW competes directly for spectral weight with the  $d$ -symmetry superconductor at the  $\mathbf{k}$ -space ‘hot frontier’ between superconductivity and the pseudogap.



## Figure Captions

### **Figure 1 *d*-symmetry form factor density wave in cuprate pseudogap phase.**

- A. Schematic phase diagram of hole-doped cuprates. The pseudogap regime has been identified by, for example, suppression of uniform magnetic susceptibility and electronic specific heat, and the appearance of a truncated Fermi surface referred to as the 'Fermi Arc' <sup>1,2</sup>. The dome-shaped region of *d*-symmetry Cooper paired high temperature superconductivity is universally accepted. More recently, an unusual density wave state has been detected by bulk probes <sup>4-13</sup> in the region indicated schematically in pink; its modulations are now known to have a *d*-symmetry form factor <sup>14-17</sup>. The range of hole-density,  $p$ , in which *d*-symmetry form factor density waves are studied in this paper is indicated by the white double-headed arrow.
- B. Schematic of the electronic structure in a cuprate dFF-DW. Grey dots represent the Cu sites and correspond to the white dots in 1C. The  $O_x$  and  $O_y$  sites within each  $\text{CuO}_2$  unit-cell are electronically inequivalent as represented by a color scale ranging from yellow through white to blue. The schematic DW modulates horizontally with wavelength  $\lambda$  or with wavevector  $\mathbf{Q}_x$  (horizontally) and with period  $4a_0$ . The periodic modulations at  $O_x$  sites are  $\pi$  out of phase with those at  $O_y$  sites as seen by considering the two trajectories marked  $\phi_x$  and  $\phi_y$  (SI Section 1).
- C. Measured  $Z(\mathbf{r}, 150\text{meV})$  at  $T > T_c$  in the pseudogap phase of  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  at hole-density  $p \sim 8\%$ . Two periods of dFF-DW modulation at  $\mathbf{Q}_x$  that correspond directly to the schematic in 1B are shown. Thus, to observe the dFF-DW state sublattice-phase-resolved imaging is required and achieved here in the pseudogap regime. The transparent overlay shows the relationship between locations of Cu,  $O_x$ ,  $O_y$  atoms in the  $\text{CuO}_2$  plane and the dFF-DW whose wavevector here is along the x-direction

**Figure 2 dFF-DW domains in superconducting and pseudogap phases.**

- A. Measured  $Z(\mathbf{r}, 150\text{meV})$  at  $T \ll T_c$  in the superconducting phase of hole density  $p \sim 8\%$  doped  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$  ( $T \sim 4.2\text{K}$ ). These complex spatial features involve modulations that comprehensively maintain a relative phase of  $\pi$  between  $O_x$  and  $O_y$  in a disordered  $d$ -symmetry FF density wave.
- B. Measured  $Z(\mathbf{r}, 150\text{meV})$  at  $T > T_c$  in the pseudogap phase of hole density  $p \sim 8\%$  doped  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  ( $T \sim 45\text{K}$ ). Although correlation lengths are shorter, the dFF-DW phenomena are otherwise indistinguishable from observations at  $T \ll T_c$ .
- C. The  $d$ -symmetry form factor power spectral density  $|\tilde{D}^Z(\mathbf{q})|^2 = |(\tilde{O}_x(\mathbf{q}) - \tilde{O}_y(\mathbf{q}))/2|^2$  determined from sublattice-phase resolved analysis of data in A. Two primary DW peaks at  $\mathbf{Q}_x$  and  $\mathbf{Q}_y$  exist with this  $d$ -symmetry form factor, as identified by dashed circles.
- D. The  $d$ -symmetry FF power spectral density  $|\tilde{D}^Z(\mathbf{q})|^2 = |(\tilde{O}_x(\mathbf{q}) - \tilde{O}_y(\mathbf{q}))/2|^2$  determined from sublattice-phase resolved analysis of data in B. Again, two primary DW peaks at  $\mathbf{Q}_x$  and  $\mathbf{Q}_y$  exist with this  $d$ -symmetry form factor, showing that the  $\mathbf{q}$ -space structure of dFF-DW phenomenology is identical in the pseudogap phase and in the superconducting phase.
- E. F. Using only the regions within the dashed circles in C,D, the  $\mathbf{r}$ -space amplitudes of the dFF-DW in A,B are calculated for modulations along  $\mathbf{Q}_x$  from Eqn. 2a, and along  $\mathbf{Q}_y$  from Eqn. 2b. Then using  $F(\mathbf{r}) = (|D_x(\mathbf{r})| - |D_y(\mathbf{r})|)/(|D_x(\mathbf{r})| + |D_y(\mathbf{r})|)$  (see SI Section 3) regions primarily modulating along  $y$ -axis with  $-1.0 < F(\mathbf{r}) < -0.3$  are shaded blue. Regions primarily modulating along  $x$ -axis with  $+0.3 < F(\mathbf{r}) < +1.0$  are shaded orange.
- F. Domain configuration of unidirectional dFF-DW modulations contained in Fig. 1A at  $T \ll T_c$ . The unidirectionality color scale for  $F(\mathbf{r})$  demonstrated in E,F is overlaid upon the data in A. The dashed circle shows the  $\mathbf{r}$ -space radius equivalent to the  $\mathbf{q}$ -space filter used to generate the  $D_{x,y}(\mathbf{r})$  images by Fourier filtering (see SI Section 4).

- G. Domain configuration of unidirectional dFF-DW modulations contained in Fig. 1B at  $T > T_c$ . The unidirectionality color scale for  $F(\mathbf{r})$  demonstrated in E,F is overlaid upon the data in B. The dashed circle has same definition as in G.

### Figure 3 Concentration of dFF-DW Spectral Weight on Pseudogap Energy

- A. The tunneling density of states spectrum  $g(E=eV)=dI/dV(E)$  typical of underdoped cuprates show for the  $p \sim 8\%$  samples presented in this paper. The energy  $\Delta_0$  beyond which Bogoliubov QPI do not exist<sup>3233</sup> and the pseudogap energy  $\Delta_1$  are indicated.
- B. The energy dispersion of seven dispersive modulation characteristic of Bogoliubov quasiparticle excitations of a  $d$ -wave superconductor ( $\mathbf{q}_1 \dots \mathbf{q}_7$ ). These Bogoliubov quasiparticle interference modulations are all simultaneously observable only below the energy  $\Delta_0$  as indicated by dashed red line (Refs.31, 32, 33); here we demonstrate that they exhibit a predominantly  $s'$ -symmetry form factor indicated by the red color. At energies above  $\Delta_0$ , the electronic structure images evolve quickly to consist of only non-dispersive  $\mathbf{Q}_x^d$  and  $\mathbf{Q}_y^d$  wavevectors of the  $d$ -symmetry form factor DW. We plot the dispersion of these modulations as the energy dependence of the maxima in  $D(\mathbf{q}, E)$  using blue squares. The same physical modulations when analyzed using  $S'(\mathbf{q}, E)$  appear as the non-dispersive  $\mathbf{Q}_x^{s'}$  and  $\mathbf{Q}_y^{s'}$  wavevectors shown as red circles.
- C. Measured  $|\tilde{D}^Z(\mathbf{q}, 88 \text{ meV})|^2$  for samples studied herein. The  $\mathbf{Q}_x^d$  and  $\mathbf{Q}_y^d$  wavevectors of the  $d$ -symmetry form factor DW are indicated by two red circles; the data of relevance for determining energy/momentum dependence of the dFF DW modulations is contained within.
- D. Measured doping dependence of  $\mathbf{Q}_x$  and  $\mathbf{Q}_y$  of  $d$ -symmetry form factor DW in underdoped  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$  is shown using blue symbols. The measured doping dependence of  $\mathbf{q}$ -vector linking tips of arcs beyond which the signature of Bogoliubov quasiparticles disappears (E & Refs.31,32,33) shown by all other colors.

- E. Measured doping dependence of the wavevectors interconnecting the  $\mathbf{k}$ -space arc tips at which Bogoliubov quasiparticle signatures disappear (B & Refs.31,32,33).
- F. Measured energy dependence of  $S'(E)$  where  $\mathbf{q}$  is integrated over the region inside solid red circles in C, is shown in red. Measured energy dependence of  $S(E)$  where  $\mathbf{q}$  is integrated over the region inside solid red circles in C, is shown in black. Measured energy dependence of  $d$ -symmetry form factor  $D(E)$  where  $\mathbf{q}$  is integrated over the region inside solid red circles in C is shown in blue (SI Section 4). These data reveal that the dFF-DW spectral weight is concentrated at energy surrounding  $\sim 90\text{meV}$  which, at this hole density, is the independently measured pseudogap energy scale  $\Delta_1$  (see Fig. 3A) and indicated on all relevant panels by a dashed blue line.

**Figure 4 Relationship between dFF-DW modulations of filled and empty states**

- A. Differential tunneling conductance image  $g(\mathbf{r}, +94\text{meV})$  measured above  $E_F$  near the pseudogap energy  $+\Delta_1$ . The color scale is reversed compared to B.
- B. Differential tunneling conductance image  $g(\mathbf{r}, -94\text{meV})$  measured below  $E_F$  near the pseudogap energy  $-\Delta_1$ . The color scale is reversed compared to A.
- C. The spatial phase of the dFF-DW modulating along the  $y$ -direction  $\phi_y(\mathbf{r}, E)$  is calculated using Eqn. 4a from  $g(\mathbf{r}, +94\text{meV})$  data in A. The dashed circle shows the  $r$ -space radius equivalent to the  $q$ -space filter used to generate the  $\phi_{x,y}(\mathbf{r}, E)$  images by Fourier filtering (see SI Section 6).
- D. The spatial phase of the dFF-DW modulating along the  $y$ -direction  $\phi_y(\mathbf{r}, E)$  is calculated using Eqn. 4b from  $g(\mathbf{r}, -94\text{meV})$  data in B.
- E. & F. From the FOV of A and B, we show the energy dependence of the relative phase of  $g(\mathbf{r}, -E)$  and  $g(\mathbf{r}, +E)$  modulations along the  $y$ -axis (SI Section 5):  $\phi_y(\mathbf{r}, +E) - \phi_y(\mathbf{r}, -E)$  when averaged over every pair of identical pixel locations  $\mathbf{r}$ , similarly for

relative phase of  $g(\mathbf{r}, -E)$  and  $g(\mathbf{r}, +E)$  for modulations along the  $x$ -axis:  $\phi_x(\mathbf{r}, +E) - \phi_x(\mathbf{r}, -E)$ . The low energy  $E < \Delta_0$  Bogoliubov quasiparticle modulations at  $+E$  and  $-E$  are in phase spatially and so have relative phase difference of 0. As the pseudogap energy  $\Delta_1$  is approached and the dFF-DW phenomena emerge, the relative spatial phase of empty-state and filled state dFF-DW modulations varies wildly in the narrow energy range shaded gray, and then quickly develops a robust phase shift of  $\pi$ .

## Acknowledgements

We acknowledge and thank H. Alloul, D. Chowdhury, R. Comin, A. Damascelli, E. Fradkin, D. Hawthorn, J. E. Hoffman, M.-H. Julien, D. H. Lee, M. Norman, and C. Pepin for helpful discussions and communications. We are especially grateful to S.A. Kivelson for key scientific discussions and advice. Experimental studies were supported by the Center for Emergent Superconductivity, an Energy Frontier Research Center, headquartered at Brookhaven National Laboratory and funded by the U.S. Department of Energy under DE-2009-BNL-PM015, as well as by a Grant-in-Aid for Scientific Research from the Ministry of Science and Education (Japan) and the Global Centers of Excellence Program for Japan Society for the Promotion of Science. CKK acknowledges support under *FlucTeam* program at Brookhaven National Laboratory Contract DE-AC02-98CH10886. S.D.E., J.C.D. and A.P.M acknowledge the support of EPSRC through the Programme Grant ‘*Topological Protection and Non-Equilibrium States in Correlated Electron Systems*’. Theoretical studies at Cornell University were supported by NSF Grant DMR-0955822. Theoretical studies at Harvard University were supported by NSF Grant DMR-1103860 and by the Templeton Foundation. Research at Perimeter Institute is supported by the Government of Canada through Industry Canada and by the Province of Ontario through the Ministry of Research and Innovation.

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Figure 1

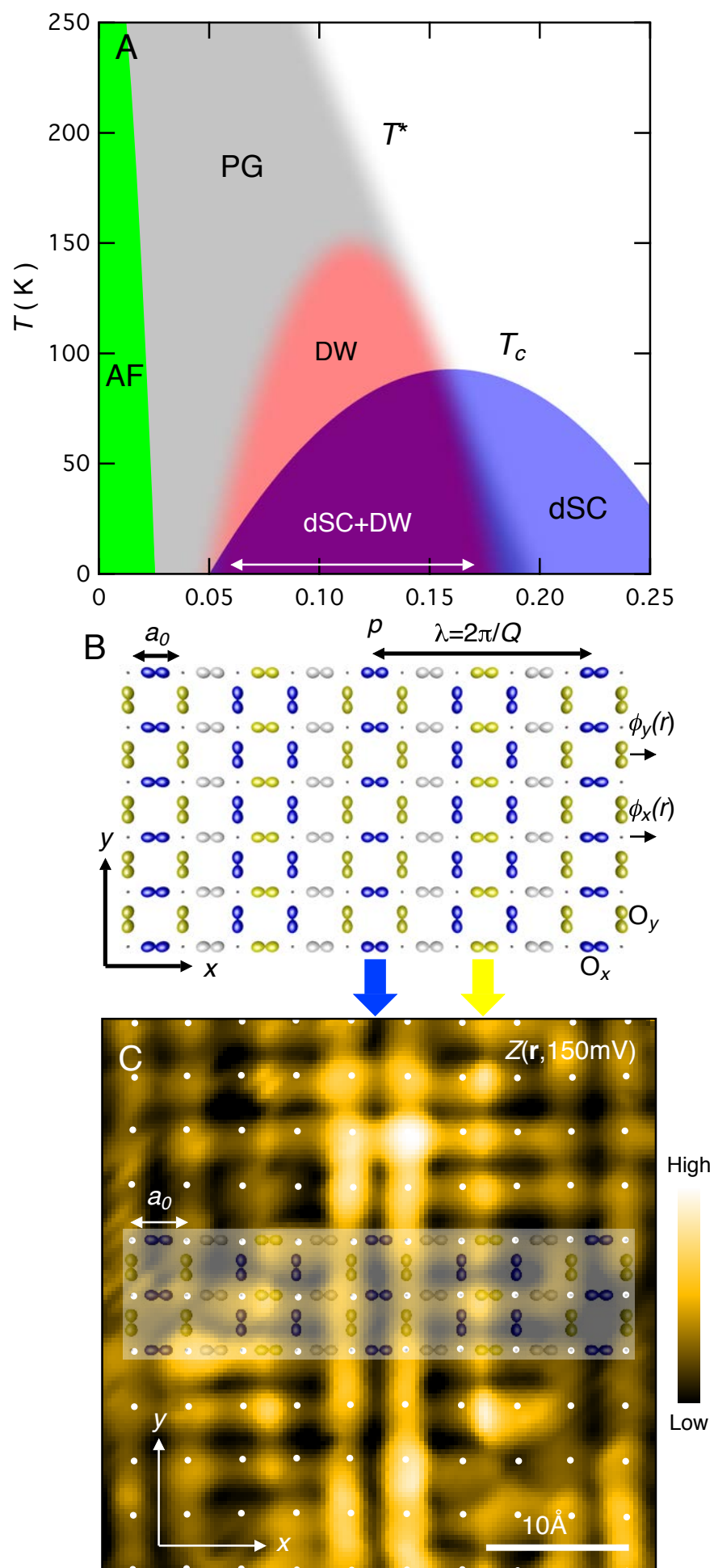


Figure 2

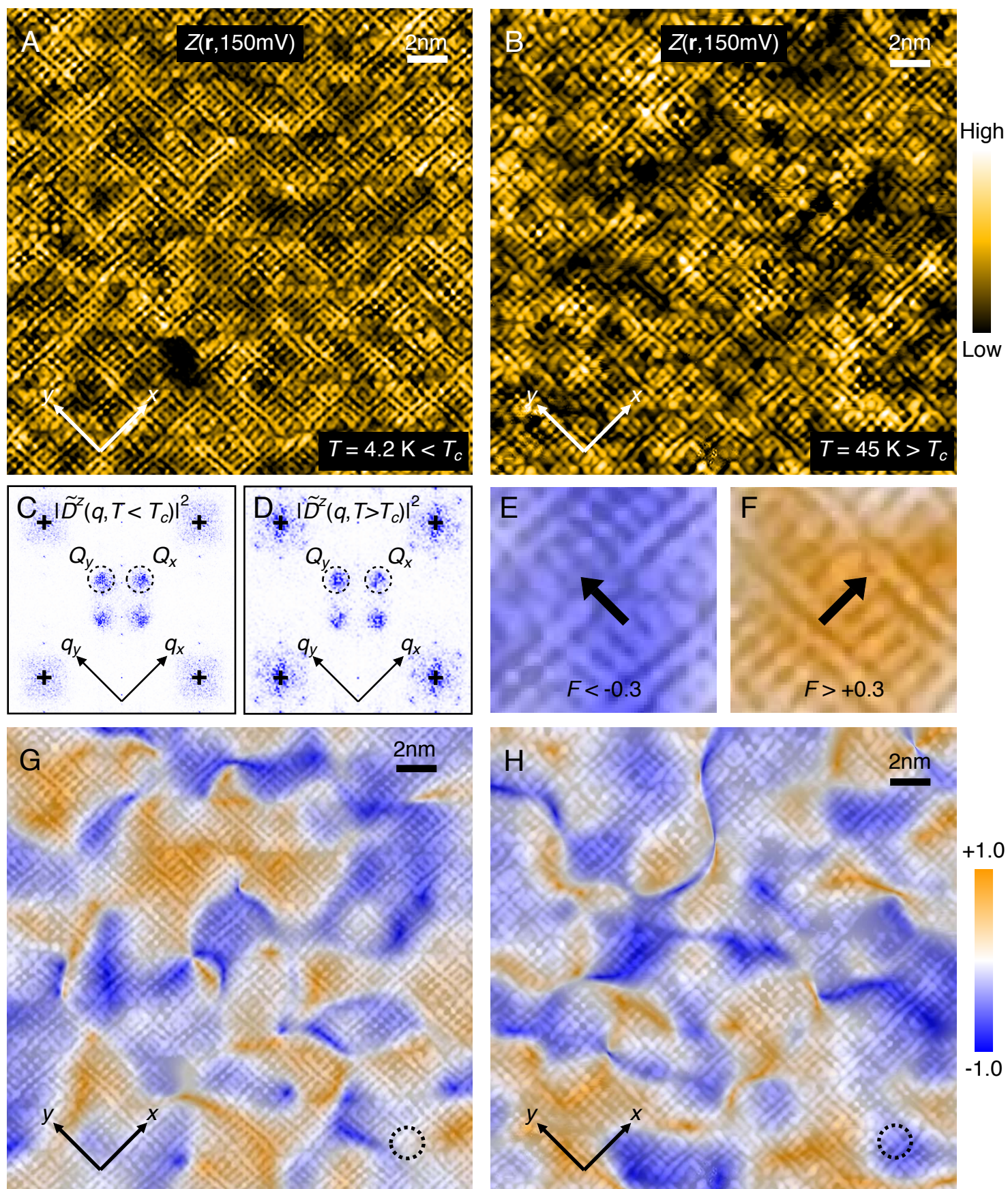




Figure 3

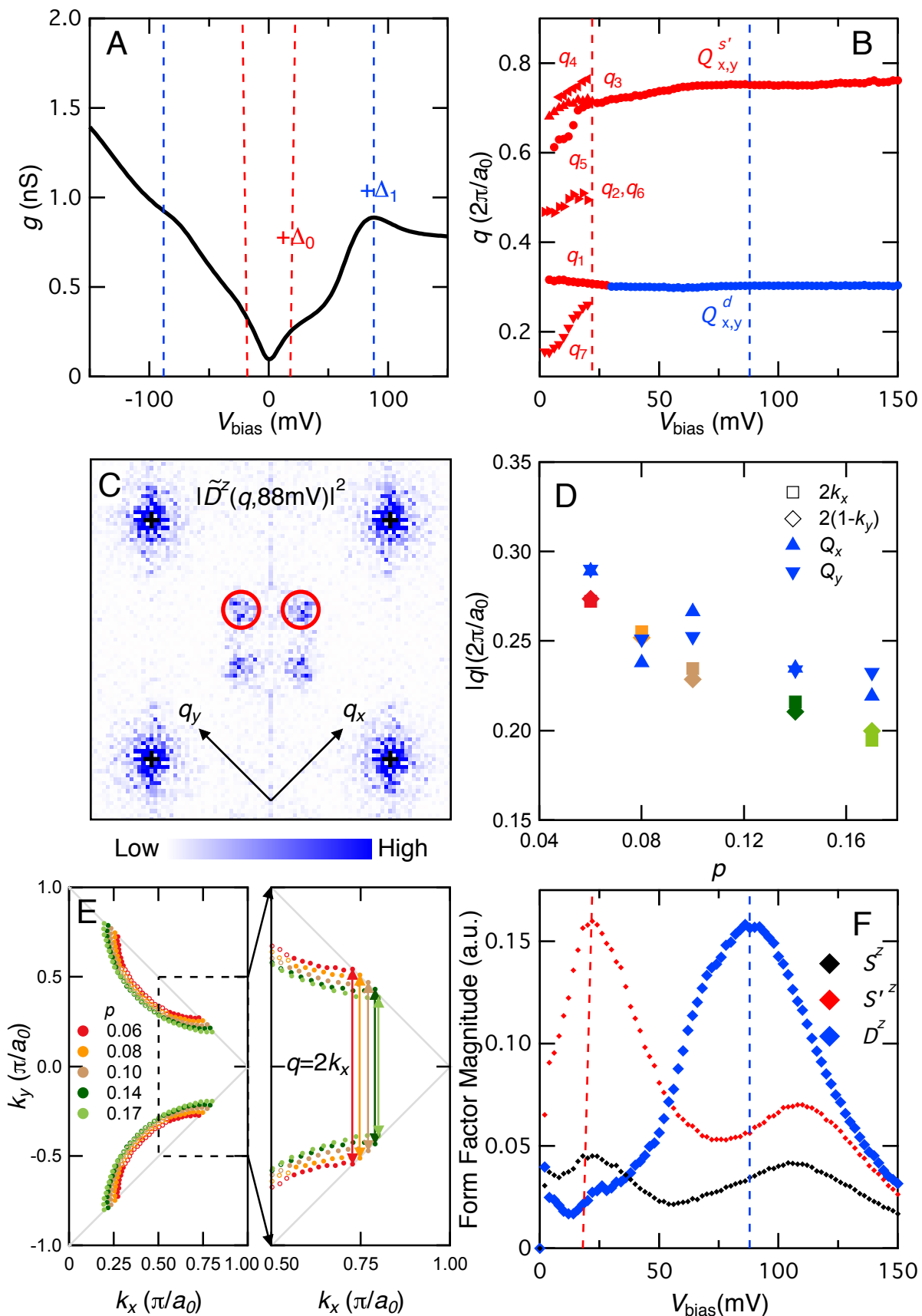


Figure 4

